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**CHARACTERIZATION OF URANIUM CONTAMINATED SOILS FROM DOE
FERNALD ENVIRONMENTAL MANAGMENT PROJECT SITE: RESULTS
OF PHASE I CHARACTERIZATION, OR NATIONAL LABORATORY -
(USED AS A REFERENCE IN OU5 RI AND FS REPORT)**

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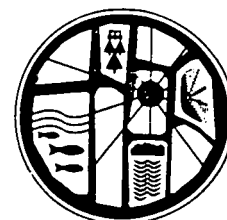


**Characterization of Uranium
Contaminated Soils from
DOE Fernald Environmental
Management Project Site:
Results of Phase I Characterization**

S. Y. Lee
J. D. Marsh, Jr

Environmental Sciences Division
Publication No. 3786

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FOR THE UNITED STATES
DEPARTMENT OF ENERGY



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ENVIRONMENTAL SCIENCES DIVISION

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FROM DOE FERNALD ENVIRONMENTAL MANAGEMENT
PROJECT SITE: RESULTS OF PHASE I CHARACTERIZATION**

S. Y. Lee
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ABSTRACT

LEE, S. Y. and J. D. MARSH, Jr. 1992. Characterization of uranium contaminated soils from DOE Fernald Environmental Management Project Site: Results of Phase I characterization. ORNL/TM-11980. Oak Ridge National Laboratory, Oak Ridge, Tennessee. 70 pp.

The Integrated Demonstration (ID) for remediation of uranium-contaminated soils has been established by the DOE Office of Technology Development. The Fernald site was selected as the DOE facility for the field demonstration. The principle objective of this ID is to evaluate and compare the versatility, efficiency, and economics of various technologies that may be combined into systems for the removal of uranium from contaminated soils.

The ID Characterization Task group designed a study to obtain basic information relating to soil properties and the nature of uranium contamination at the site soil. Such information is essential for the selection of (1) contaminated soils for use in treatability studies, (2) a field demonstration area at the site, and (3) integrated technologies. The task group selected five areas and collected two core samples from each area.

The nature of soil contamination was investigated by examining (1) uranium distribution with soil depth, (2) soil particle size distributions and their uranium contribution, (3) soil chemical and physical properties, (4) particle density of soil and contaminant, (5) mineralogical and microscopic properties of soil and contaminant, (5) chemical leaching characteristics, and (6) background soil uranium content and soil properties.

The results indicated: (1) except in an area contaminated by acidic solution spills, the contamination depth of most areas was shallow (usually <10 cm containing from 10 to 2800 pCi/g); (2) background uranium concentration of off-site soils was <4 pCi/g; (3) the sand and silt size fraction of soils contained from 48 to 79% of the uranium in soils; (4) the dominant form of uranium was sand and silt-sized particulate often associated with

calcium, phosphorous, iron, and silicon; (5) most of the uranium particulates had a density >2.9 g/mL, however; and (6) considerable amounts of soil uranium, 10 to 49% and 20 to 75%, could be extracted using 2% solutions of ammonium carbonate and citric acid, respectively.

On the basis of the soil characterization results, two areas, Plant 1 Drum Storage area and the Incinerator area, were selected for use in treatability studies and insitu characterization demonstration areas. The Plant 1 Drum Storage area was contaminated by uranium product spills and the Incinerator area was contaminated by airborne uranium materials during incineration of contaminated materials. Particulate uranium was the dominant form associated with the sand and silt fractions of both soils (see attached micrograph plate). Some uranium in the soils was not readily extractable. Therefore, simple chemical extraction alone would not be effective for waste volume reduction as soil remediation. Development of more effective and selective extraction technology and density-based physical separation technology is needed to meet the cleanup goal for the Integrated Demonstration.

**CHARACTERIZATION OF URANIUM-CONTAMINATED SOILS
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RESULTS OF PHASE I CHARACTERIZATION**

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INTRODUCTION

The Integrated Demonstration (ID) for uranium-contaminated soils remediation has been established by the DOE Office of Technology Development. The principle objective of this ID is to evaluate and compare the versatility, efficiency, and economics of various technologies that may be combined into systems for the removal of uranium from contaminated soils. Because the scope of the ID program is to address remedial alternatives for uranium-contaminated soils, the Fernald Site was selected as the DOE facility for the field demonstration. The draft RI/FS report (DOE, 1990) of the site concluded that the majority of uranium-contaminated soils were located within the Operable Unit 3, including the Sewage Treatment Plant/Old Incinerator area (Figs. 1 and 2). Therefore, the ID team selected Operable Unit 3 as the source of contaminated soils for the field demonstration (Note: After the renegotiation of the CERCLA Consent Agreement, the management of all soils became the responsibility of Operable Unit 5.)

The Phase I soil sampling/characterization task was established by the Characterization Group to obtain basic information related to soil properties and the nature of uranium contamination. Such information is essential for:

- the selection of contaminated soils for use in treatability studies,
- the selection of a field demonstration area or areas within Operable Unit 3, and
- the preliminary screening of integrated technologies (Tidwell's Memorandum, 1991).

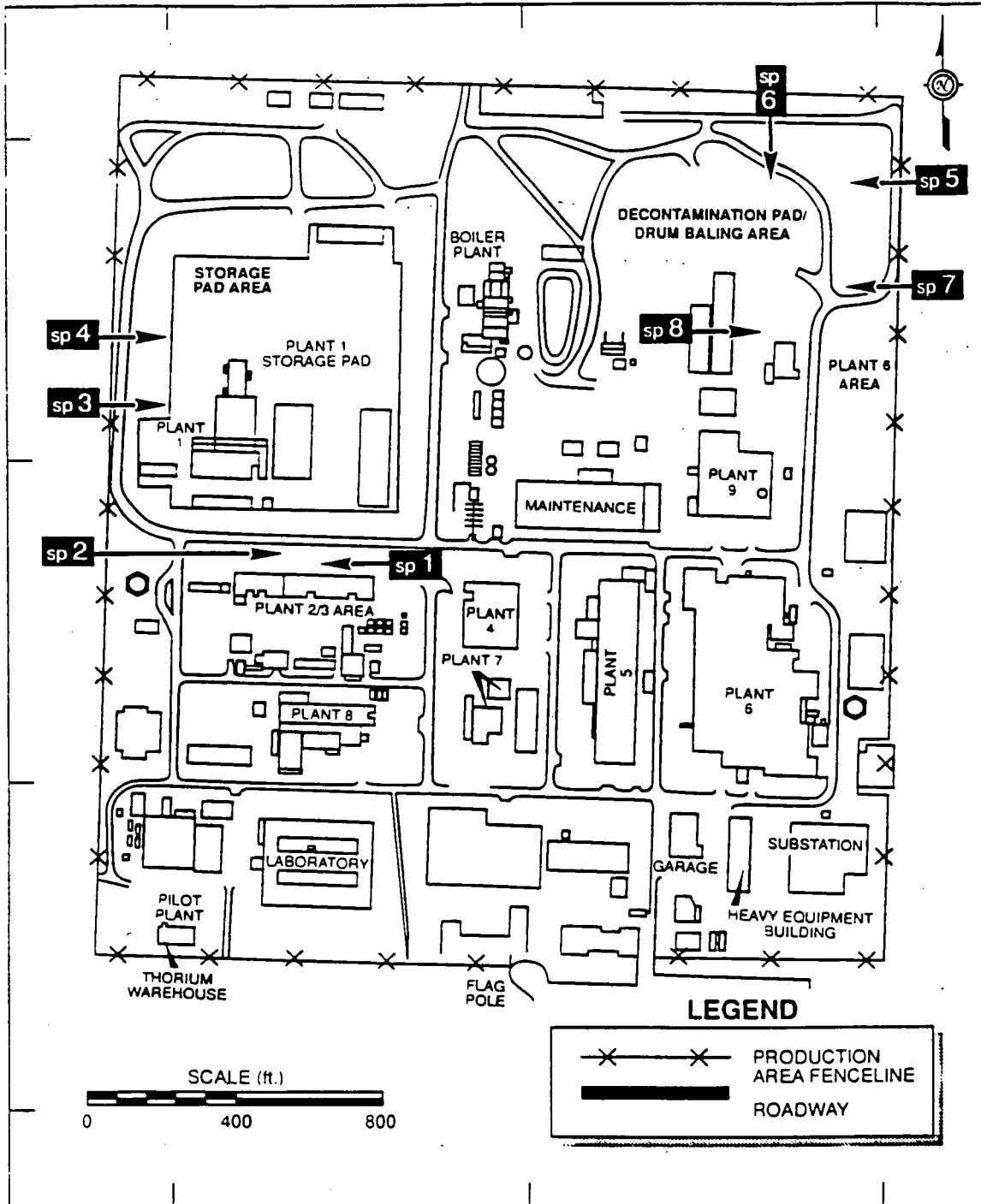


Figure 1. Sampling locations in Operable Unit 3 of Fernald Operation Site.

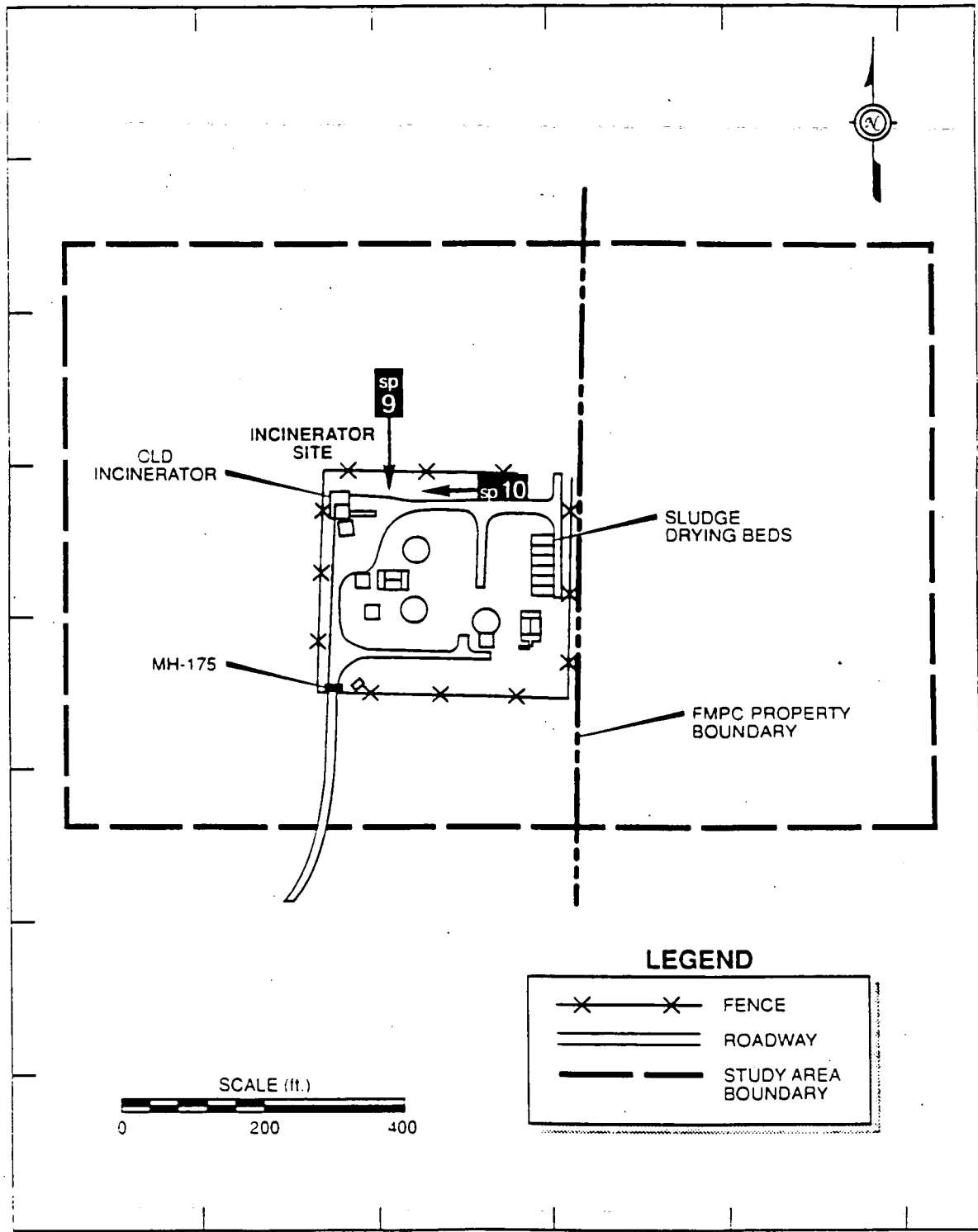


Figure 2. Sampling locations in the incinerator area of Fernald Operation Site.

Table 1 is a list of the information needs. More detailed characterization will be conducted in the Phase II investigation that will follow the Phase I investigation. This Phase I investigation is intended to be finished within 2 1/2 months because of the schedule established by DOE for preliminary treatability tests at Nevada.

This preliminary investigation will, however, provide critical information related to: (1) the nature of uranium contamination, (2) important soil properties related to uranium retention, (3) guide the direction on the next phase of the investigation, and (4) narrow the scope of technology investigations.

Table 1. Characterization information needs for the Integrated Demonstration Program

Properties	Where the information is needed
1. Uranium distribution with depth:	Excavation, Risk assessment
2. Soil particle size distribution:	Treatability, Risk assessment, Waste Disposal
3. Uranium distribution with particle size:	Treatability, Risk Assessment
4. Soil chemical and physical properties:	Excavation, Treatability
5. Specific gravity soil/contaminant:	Treatability
6. Soil solution chemistry:	Treatability, Risk Assessment, Site Operation
7. Mineralogical analysis:	Treatability, Waste Disposal
8. Microscopic analysis:	Treatability
9. Uranium form identification:	Treatability, Risk Assessment
10. Chemical leaching test:	Treatability, Waste Disposal, Risk Assessment
11. Reference soil characterization:	Risk Assessment, Treatability, Regulation

Characterization Group members agreed to select soil sampling areas based on the contamination source term characteristics, that is, aqueous uranium wastes, solid uranium product spills, and airborne uranium wastes (dust, aerosols) (Tidwell's Memorandum, 1991). The decontamination Pad/Drum Baling area and north of the Plant 2/3 area have

been selected to represent the soils contaminated by aqueous uranium wastes. The Plant 1 Drum Storage area was selected for the soils contaminated by uranium product spills. And, the Incinerator and Plant 6 areas were selected to represent the soils contaminated by airborne releases of uranium. Two reference soils, Fincastle and Henshaw Series located about 1 mile and 1.5 miles west of the Fernald Site, respectively, were collected for base line data establishment (Fig. 3).

METHODS

Collection of soil core samples for the Phase I Sampling Program was performed according to the protocols and procedures established for the RI/FS Operable Unit 3 program, the RI/FS Quality Assurance Project Plan, and the Project Specific Health and Safety Plan (Tidwell's Memorandum, 1991).

Characterization Group representatives (S. Y. Lee and Mark Nicholson) with the aid of the Fernald RI/FS sampling team and Health Physics personnel performed site surveys with a sodium-iodide survey meter at selected sampling sites. Specific sampling points were selected according to areas exhibiting a high activity. Prior to collecting sample, gravel or grass covers were removed before setting up a hand-driven auger. A stainless steel auger with one or three 12-in.-long polybutyrate sleeve (2 in. diam) was used for sample collection. After retrieval of the auger by a hydraulic jack, soil cores were cut at the joints of sleeves and capped for shipment. Supplemental undisturbed samples were collected by pushing down an 8.5-cm-diam \times 3.5-cm-deep plastic dish and cutting the bottom of the soil block with a knife for microscopic analyses. Samples were shipped to the Oak Ridge National Laboratory for characterization.

The soil cores were described according to the standard soil description methodology (Soil Survey Staff, 1975) and then cut into 2- to 4-in. segments. Each soil core segment was given an identification number in the following way; SP#-1-A, "SP#" representing sampling site, "-1" representing the order of sleeve from the top or the order of sampling when three 12-in. cores were taken instead of one 36 in. core per each sampling location, and "-A" representing the order of each soil segment starting from the top of each sleeve.

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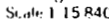


Figure 3. Reference soil sampling locations on soil survey map of Hamilton County, Ohio.
(FdA = Fincastle Series; HoA = Henshaw Series)

Soil pH measurements were made using a PHM 84 Research pH meter with a combination pH electrode. The meter and electrode were calibrated with standard pH buffers of pH 7 and 10 to encompass the pH range of the soil samples. The soil samples were prepared by adding 5 ml of distilled water to 5 g of soil, stirring, and allowing the soil and water to set in contact for about 6 h before beginning pH measurements. The electrode was left in the soil/water mixture until the reading had stabilized and the measurement recorded.

The soil core segments (3 to 4 in.) were transferred to 8.5-cm-diam (internal) by 3-cm-high plastic containers for gamma spectroscopy. Gamma analysis was done on all the samples prior to particle size separation. The samples were counted on a high resolution, solid state, coaxial, intrinsic, germanium (IG) detector coupled to an ND6700 multichannel analyzer with 4096 channels. The gamma system had previously been calibrated with a Amershan QCY44 certified mixed gamma standard with traceability to National Institute of Standards and Technology (NIST), formerly the National Bureau of Standards (NBS), in the geometry used to contain the soil samples. In addition, weekly and daily sources were counted to verify that the detectors were remained calibration. Samples were counted down the soil column from 1 h to overnight depending on their level of activity. The ^{238}U concentration was determined using the 1001-keV line for ^{234}Pa , which is a daughter of ^{238}U assumed to be in secular equilibrium with the uranium isotope. At equilibrium the activities of the two nuclides should be the same and the measurement of one determines the activity of the other. The ^{235}U was determined using its 143-keV peak. The activity ratio of $^{235}\text{U}/^{238}\text{U}$ for natural uranium is 4.6. Cesium-137 at 661 keV and ^{40}K at 1460 keV were also measured down the soil column. For those samples with activities below detectable levels, a minimum detectable activity (MDA) is reported. This is the minimum activity of the radionuclide which have to be present 95% of the time to be detected in the presence of the sample compton continuum.

After gamma spectroscopy, soil segments from the SP22, SP2-3, SP4, SP5, SP8, and SP9 cores were selected. The selected segments from each core were combined as needed to obtain enough sample for characterization. For example, a soil sample identification number such as SP2-2-ABC represents a mixed sample of A, B, and C segments from the

SP2-2 sleeve and SP4-1A/2A represents a mixed soil of the first segment of the first sleeve and the first segment of the second sleeve from SP4.

Particle size separation of the selected soil samples was performed by dry sieving with 4- and 2-mm sieves (size fractions larger than 2 mm were designated as gravel). The <2-mm fractions were further separated into 2 to 0.053 mm (sand), 0.053 to 0.002 mm (silt), and <0.002 mm (clay) by wet sieving and centrifugation method (Jackson 1975). Water samples produced during particle-size separation and soil samples were submitted to the Environmental Analysis Laboratory located at the Y-12 Plant in Oak Ridge, Tennessee, for the following analyses; total uranium by mass spectroscopy, isotopic uranium by alpha spectroscopy, trace element analysis by inductively coupled plasma atomic emission spectroscopy (ICP-AES), anions by ion chromatography (IC), and alkalinity by acid titration to pH 4.5. Due to the sensitivity of the alpha spectroscopy method for uranium, only those soils and leachates with very low levels of uranium (<10 pCi/g for solid and <100 pCi/L for liquid) could be analyzed by this method. For this reason, the high-level uranium samples were done by mass spectroscopy. Prior to the uranium and metal analyses, 1 to 3 g of soil was digested at 90 to 95°C with nitric acid and 30% hydrogen peroxide, centrifuged, and filtered. The filtrates were diluted before spectroscopy analyses.

Several leaching solutions were employed to determine their effectiveness in extracting uranium from the soil. The extractants and their means of preparation were:

- 0.1 N nitric acid [HNO_3]: 6.25 mL of concentrated nitric acid was diluted to 1 L with distilled water.
- 2% ammonium carbonate [$(\text{NH}_4)_2\text{CO}_3$]: 20 g of $(\text{NH}_4)_2\text{CO}_3$ was dissolved in distilled water and diluted to 1 L.
- 5% sodium hypochlorite (NaOCl): 50 mL of NaOCl reagent (Cl < 6%) was diluted to 1 L with distilled water.
- 0.1 M ethylenediaminetetraacetic acid, disodium salt (EDTA): 37.224 g of EDTA was dissolved in distilled water and diluted to 1 L.

- 2% citric acid monohydrate solution ($\text{H}_3\text{C}_6\text{H}_5\text{O}_7\cdot\text{H}_2\text{O}$): 20 g of citric acid was diluted to 1 L with distilled water.
- 0.1 M hydroxylamine-hydrochloride ($\text{NH}_2\text{OH}\cdot\text{HCl}$) in 0.01 N nitric acid: 6.95 g ($\text{NH}_2\text{OH}\cdot\text{HCl}$) was dissolved and diluted to 1 L with 0.01 N HNO_3 . The 0.01 N nitric acid was prepared by diluting 3 mL concentrated nitric acid to 5 L with distilled water.

The procedure for each extraction was the same except for the extractant used. The soils extracted were ones that had been sieved and consisted of particles <2 mm in size. Those samples were: SP2-2-ABC, SP2-3-ABC, SP4-1A/2A, SP5-1-AB, SP8-1A/2A/3A, and SP9-1A/3A. Forty milliliters of the extractant were added to 5 g of each soil (1:8 soil/solution ratio) and mixed for 2 h in a shaker. The samples were then centrifuged for 6 min at about 3000 rpm in an IEC HN-SII centrifuge. The liquid was decanted and filtered through a 0.45- μm , 25-mm, Acrodisc. This leachate was then submitted to the Y-12 Environmental Analysis Laboratory for total uranium analysis by mass spectroscopy and trace element analysis by inductively coupled plasma atomic emission spectroscopy (ICP-AES). In addition to the leachates, some unleached soil was also submitted for analysis so that the percent of uranium and trace metals extracted could be calculated.

One additional extraction was conducted employing the citrate-bicarbonate-dithionite (CBD) method. This method is summarized as follows.

- Sodium citrate-bicarbonate-dithionite (CBD) method: 0.3 M sodium citrate (88 g tribasic sodium citrate, $\text{Na}_3\text{C}_6\text{H}_5\text{O}_7\cdot 2\text{H}_2\text{O}$, per liter); 1 M sodium bicarbonate (84 g NaHCO_3 per liter); and 5 g sodium dithionite, $\text{Na}_2\text{S}_2\text{O}_4$.

For the CBD extraction, 800 mL of sodium citrate were mixed with 100 mL sodium bicarbonate for an 8:1 citrate/bicarbonate solution. Sixty milliliters of this solution were added to 15 g of soil in a 200-mL centrifuge bottle. The soil plus citrate/bicarbonate solution was then heated in a water bath to 75-80°C. At about 78°C, 5 g of sodium dithionite were added and the mixture stirred for 15 min. After digestion, the mixture was centrifuged, the solution decanted, and filtered through a 0.45- μm , 25-mm, Acrodisc. This

leachate was also submitted to the Y-12 Environmental Analysis Laboratory for total uranium by mass spectroscopy and trace element analysis by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

The CBD treatment soil samples were separated into sand-, silt-, and clay-sized fractions by the wet sieving and centrifugation methods for mineralogical analysis by x-ray powder diffraction (XRD) and heavy liquid density separation. Clay fractions were saturated with magnesium and potassium and excess salts were removed by washing. Oriented clay specimen slides, two for magnesium-saturated clay and three for potassium-saturated clay were prepared using the filter membrane peel technique (Drever 1973). One of the magnesium-saturated clay slides was solvated with ethylene glycol and the second and third sets of potassium-saturated clay slides were heated at 300 and 550°C. XRD scans began and ended at 2 and 30 degrees, two theta, respectively, using copper K alpha radiation on a Norelco-Philips x-ray diffractometer.

The dry, undisturbed, surface soil samples collected in plastic dishes and soil clumps from the subsurface were embedded in epoxy resin under a vacuum allowing the solution to move into soil micropores. After resin polymerization, microscopic specimens about 2×3.5 cm), were prepared by cutting the soil resin blocks perpendicular to the soil surface by a diamond saw. The specimens were polished with silicon carbide powder. Scanning electron microscopy (SEM), utilizing both secondary electron imaging (SEI) and backscattered electron imaging (BEI) in conjunction with energy dispersive X-ray analysis (EDX), was used for analysis of morphology, particle size, and elemental distributions (Lee 1990).

Lithium metatungstate solution with density 2.8 g/mL was used for the heavy liquid density separation. About 10 mL of the lithium metatungstate solution was transferred into a 20-mL plastic centrifuge tube and to about 3 g of sand fractions separated after CBD treatment to the solution. After mixing for 5 min and centrifugation for 10 min (2000 rpm), the bottom of the tube containing heavy particles was placed in liquid nitrogen to freeze the bottom, thus allowing the upper (floating) part to be poured off to separate the fractions. Both the heavy and floating fractions were collected on filter

papers (0.45 μm) and washed with distilled water using a vacuum filtering apparatus. The fractions were placed in petri dishes, and uranium concentrations were determined by gamma spectroscopy as described above. A portion of the floating and heavy fractions was embedded in epoxy resin solution for SEM, EDX, and XRD analyses (in progress). These analyses would provide information related to elemental composition and crystalline phase of uranium containing particles.

RESULTS AND DISCUSSION

Soil Description and Gamma Spectroscopic Analysis

Reference Soils: Undisturbed soils on and around the Fernald Site are classified as either Fincastle or Henshaw series. The Henshaw series consists of deep, somewhat poorly drained, moderately permeable soils that formed in alluvium from calcareous loess (SCS 1982). Henshaw soils would be on a stream terrace as seen from the southeast portion of the Fernald Site. The Fincastle series consists of deep, somewhat poorly drained soils that formed in loess and in the underlying loam till. Permeability of Fincastle soils is moderate in the upper solum and moderately slow in the underlying glacial till. Current usage of the lands is farming. The results of gamma spectroscopic analysis are in Table 2. Uranium contents of the background soils were very close to the lower detection limit of the gamma spectroscopic analysis. Both mass and alpha spectroscopies were performed for reference soil uranium analysis.

Plant 2/3 Area: The area surrounding Plant 2/3 has been highly disturbed from past construction and decontamination activities. Limestone gravels were on the surface and mixed into the soil as deep as 30 in. from the surface. Two soil core samples, SP1 and SP2, were taken from the area (Fig. 1). Both core samples had a very high gravel content, and bulk density of the soils ranged from 1.2 to 1.9 g/cm^3 depending on the clay and moisture contents of particular core segments. Soil ranged from light yellowish brown (10YR 4/2) to yellowish gray (2.5YR 5/0). The SP1 core had a clay layer at the bottom (18+ in.) and had a very low uranium content (Table 3). The SP2 core was selected for the characterization study because the soil was contaminated uniformly down to 32 in.

Table 2. Radionuclide concentrations in reference soils at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
Fincastle Soil											
FS-1	0-6	2.56E+00	30.10	8.20E-02	39.95	3.20	1.60	1.24E+01	1.53	1.13E-01	6.54
FS-2	6-12	9.94E-01	MDA	5.16E-02	MDA			1.14E+01	1.20	9.52E-03	MDA
FS-3	12-18	1.53E+00	35.76	3.71E-02	67.84	2.42	1.85	1.46E+01	1.17	3.57E-03	122.17
FS-4	18-24	1.46E+00	55.81	1.62E-01	33.67	11.10	7.23	1.52E+01	1.90	1.67E-02	MDA
FS-5	24-30	2.70E+00	41.47	7.62E-02	51.82	2.82	1.87	1.58E+01	1.45	1.41E-02	MDA
FS-6	30-36	5.74E-01	90.49	7.71E-02	37.48	13.44	13.16	1.35E+01	1.21	9.33E-03	MDA
Henshaw Soil											
HS-1	0-6	2.50E+00	24.71	6.27E-02	51.46	2.51	1.43	1.24E+01	1.60	9.76E-02	6.89
HS-2	6-12	1.69E+00	55.24	3.12E-02	146.55	1.85	2.90	1.28E+01	2.05	2.71E-02	51.69
HS-3	12-18	1.77E+00	54.73	3.51E-02	98.65	1.98	2.23	1.24E+01	2.27	1.57E-02	MDA
HS-4	18-24	9.57E-01	MDA	4.19E-02	55.90			1.41E+01	1.00	7.16E-03	MDA
HS-5	24-30	2.24E+00	60.92	4.23E-02	65.59	1.89	1.69	1.62E+01	1.86	1.52E-02	MDA
HS-6	30-36	1.06E+00	59.76	5.57E-02	56.49	5.24	4.31	1.71E+01	1.34	1.08E-02	MDA

^a²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

^b²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

^c⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

^d¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

Table 3. Radionuclide concentrations in SP1 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP1-1-A	0-4	5.07E+02	2.53	3.27E+01	2.35	6.44	0.222	1.73E+00	28.11	2.48E-01	24.61
SP1-1-B	4-7	1.50E+03	1.68	1.05E+02	1.10	7.01	0.141	4.07E+00	19.00	3.28E-01	24.38
SP1-1-C	7-10	2.86E+01	16.52	2.25E+00	10.36	7.86	1.533	1.08E+01	6.03	4.63E-02	MDA
SP1-2-A	10-14	5.77E+02	2.42	3.73E+01	2.21	6.47	0.212	3.33E+00	12.54	1.49E-01	39.80
SP1-2-B	14-18	4.29E+02	2.55	2.96E+01	2.30	6.90	0.237	7.15E+00	8.03	1.61E-01	31.51
SP1-2-C	18-22	5.80E-00	MDA	2.43E-01	MDA	4.18	0.000	1.42E+01	5.27	4.85E-02	MDA
SP1-3-A	22-25	1.85E+01	21.67	8.22E-01	21.36	4.44	1.352	1.13E+01	6.17	4.16E-02	MDA
SP1-3-B	25-28	7.37E+00	MDA	8.25E-01	34.56	11.21	3.872	1.37E+01	5.69	4.47E-02	MDA
SP1-3-C	28-33	3.02E+00	MDA	2.09E-01	MDA	6.91	0.000	1.10E+01	6.11	3.74E-02	MDA
SP1-3-D	33-36	3.83E-00	MDA	1.85E-01	MDA	4.82	0.000	1.22E+01	4.99	2.58E-02	MDA

^a²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

^b²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

^c⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

^d¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

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Table 4 gives the activities of individual segments. In order to examine uniformity of contamination characteristics, two soil samples were chosen at the different depths from the SP2 core. The SP2-2 and SP2-3 samples were prepared by combining A, B, and C segments of the core sections from 10 to 20 in. and 20 to 31 in. depth, respectively.

Plant 1 Drum Storage Area: This area is located in the northwestern part of the plant, and the underlying soils should be Fincastle series if they have not been too deeply disturbed. Soil sampling sites are located on the west side of the Drum Storage Area. The soil sampling area was covered by fescue grass. Most of the area was mowed but the northern portion of the grass area was left alone because the area was designated as a regulated zone. The SP3 core was taken near the concrete pad in the south and the SP4 core was taken from the unmowed area in the north (Fig. 1). The unmowed area was a surface water receiving area from the Drum Storage Pad. Soil in the SP3 core had a light yellowish brown (10YR 6/4) to brown (10YR 5/3) color with weakly developed soil structure and about 30 to 60% limestone gravels. The presence of the angular limestone gravel indicated that the SP3 core area had been highly disturbed from past activities. The gamma spectroscopy results of the core indicated that the level of uranium contamination (Tables 5 and 6) was <100 pCi/g in the upper 4 in. and <10 pCi/g below 7 in. Soil in the upper part of the SP4 core had a light to dark brown color with a lower dark gray (2.5Y 4/0) clay reducing zone. Gravel content in the soil core was less than 8%. The soil had relatively abundant plant roots, loam texture in the surface horizon (Ap), and clay loam textured subsurface horizons. Depth of considerable contamination (91 pCi/g) was above 7 in. The SP3 core was selected for the Phase I investigation because of the high contamination level of the soil core.

Decontamination Pad/Drum Baling Area: This area is located in the northeast corner of the site and is used as a storage area for contaminated materials (Fig. 1). Two soil core samples were taken from the area. The SP5 core was taken from the area where the surface was covered by gravel-sized contaminated slag materials. The field survey indicated that the slag materials were the major source of the radioactivity in the area. Smaller-sized slag material was mixed into soil down to 9 in. of the core and soil had pale brown to yellowish brown color and silt loam to clay loam texture. Considerable

Table 4. Radionuclide concentrations in SP2 soil core samples at varying depths

ID	Depth (in.)	$^{238}\text{U}^a$ (pCi/g)	1 sigma % error	$^{235}\text{U}^b$ (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	$^{40}\text{K}^c$ (pCi/g)	1 sigma % error	$^{137}\text{Cs}^d$ (pCi/g)	1 sigma % error
SP2-1-A	0-4	5.41E+02	3.00	3.26E+01	2.70	6.02	0.243	5.64E+00	11.66	3.28E-01	35.71
SP2-1-B	4-6	1.07E+03	2.26	6.12E+01	2.16	5.72	0.179	5.52E+00	14.77	2.64E-01	27.92
SP2-1-C	6-10	5.84E+02	2.73	3.54E+01	2.39	6.05	0.220	5.53E+01	12.34	3.18E-01	18.90
SP2-2-A	10-13	2.10E+02	4.85	1.17E+01	6.45	5.59	0.451	6.74E+00	11.94	1.00E-01	MDA
SP2-2-B	13-16	2.37E+02	5.41	1.34E+01	4.90	5.68	0.414	7.87E+00	12.72	1.22E-01	MDA
SP2-2-C	16-20	4.75E+02	3.56	2.56E+01	3.57	5.38	0.271	5.54E+00	18.88	1.06E-01	58.25
SP2-3-A	20-24	5.75E+02	2.35	3.22E+01	2.48	5.59	0.191	4.08E+00	17.67	2.27E-01	26.06
SP2-3-B	24-27	1.83E+02	4.65	1.07E+01	5.02	5.88	0.402	1.09E+01	7.32	8.58E-02	MDA
SP2-3-C	27-31	5.29E+02	2.69	2.79E+01	2.69	5.28	0.201	6.74E+00	9.82	9.20E-02	46.61
SP2-3-D	31-32	2.63E+02	6.65	1.57E+01	5.80	5.98	0.528	1.11E+01	10.21	1.60E-01	MDA

^{238}U activity based on the 100.00-keV (0.92%) gamma energy line for $^{234\text{m}}\text{Pa}$.

^{235}U activity based on the 143.77-keV (10.50%) gamma energy line.

^{40}K activity based on the 1460.73-keV (10.70%) gamma energy line.

^{137}Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ^{235}U .
Normal atmospheric fallout from ^{137}Cs is around 1 pCi/g.

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Table 5. Radionuclide concentrations in SP3 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP3-1-A	0-4	9.08E+01	7.56	4.74E+00	8.37	5.23	0.589	9.59E+00	7.61	2.29E-01	17.70
SP3-1-B	4-7	1.67E+01	21.54	1.33E+00	14.05	7.98	2.053	1.25E+01	6.08	5.25E-02	MDA
SP3-1-C	7-10	6.40E+00	60.34	4.62E-01	45.14	7.21	5.434	1.33E+01	6.04	4.56E-02	MDA
SP3-2-A	10-13	6.88E+00	MDA	2.26E-01	MDA	3.29	0.000	1.31E+01	6.12	4.26E-02	MDA
SP3-2-B	13-16	5.82E+00	MDA	2.39E-01	MDA	4.11	0.000	1.24E+01	5.56	4.47E-02	MDA
SP3-2-C	16-18	5.33E+00	MDA	2.21E-01	MDA	4.14	0.000	1.14E+01	6.83	3.43E-02	MDA
SP3-3-A	18-21	4.60E+00	MDA	2.07E-01	MDA	4.50	0.000	9.91E+00	6.69	3.87E-02	MDA
SP3-3-B	21-24	5.98E+00	MDA	1.46E-01	84.89	2.45	2.081	1.09E+01	5.77	3.16E-02	MDA
SP3-3-C	24-27	4.76E+00	MDA	2.06E-01	MDA	4.33	0.000	9.98E+00	6.41	3.33E-02	MDA
SP3-3-D	27-30	5.80E+00	MDA	2.00E-01	MDA	3.45	0.000	1.20E+01	5.74	3.74E-02	MDA

²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

Table 6. Radionuclide concentrations in SP4 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP4-1-A	0-4	2.77E+03	1.16	1.19E+02	1.08	4.31	0.068	1.55E+01	8.59	9.77E-01	12.48
SP4-2-A	4-7	1.05E+02	6.42	3.98E+00	7.96	3.79	0.388	1.07E+01	6.53	6.79E-02	MDA
SP4-2-B	7-10	1.98E+01	16.73	9.03E-01	17.32	4.55	1.096	7.34E+00	7.19	3.55E-02	MDA
SP4-2-C	10-13	8.57E+00	37.72	1.49E-01	19.88	1.73	0.739	6.38E+00	7.76	2.65E-02	MDA
SP4-2-D	13-16	4.48E+00	52.85	4.94E-01	21.78	11.02	6.297	7.83E+00	8.13	2.88E-02	MDA
SP4-3-A	16-19	5.47E+00	MDA	2.05E-01	52.30	3.75	1.963	1.29E+01	5.61	4.58E-02	MDA
SP4-3-B	19-22	4.67E+00	MDA	1.49E-01	90.01	3.20	2.876	1.19E+01	5.85	4.39E-02	MDA
SP4-3-C	22-25	4.75E+00	MDA	1.77E-01	MDA	3.73	0.000	1.15E+01	6.24	3.80E-02	MDA
SP4-3-D	25-28	6.72E+00	MDA	2.37E-01	MDA	3.53	0.000	1.13E+01	7.03	4.34E-02	MDA

^a²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

^b²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

^c⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

^d¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

radioactivity was detected from the top 8 in. of the core. The SP6 core was taken near the railroad track and east of the Decontamination Pad (Fig. 1). The soil had yellowish brown color with clay loam texture and contained limestone fragments throughout the soil core. Gamma spectroscopy showed that uranium contamination of the SP6 soil core was relatively lower than the SP5 soil core (Tables 7 and 8). Therefore, the 0- to 9-in. segment of the SP5 soil core was chosen for characterization.

Plant 6 Area: The initial sampling plan was to collect samples from the northeast side of Plant 6. An alternate area was selected because the proposed area was disturbed by construction activities. The SP7 and SP8 core sampling sites were located further north of the initially planned area (Fig. 1). The SP7 site was considerably contaminated by waste spills or dumping activities in the area (Table 9). However, the area selected for SP8 soil core was relatively undisturbed. The area was selected to obtain a soil sample which had been contaminated by airborne uranium waste. Therefore, the SP8 soil core was investigated in this phase of characterization. The SP8 soil had light brown color, well developed soil horizons, silt loam texture, and high organic matter content contributed by growing grass in the area for considerable time. The uranium contamination was limited to only a few inches below the surface (Table 10). Therefore, a composite soil sample was prepared by combining the top 3 in. from three soil cores.

Incinerator Area: The surface soil was contaminated by the past incinerating activities of the old primitive incinerator located in the area. The contaminated area was covered by fescue grass and three 12-in. cores per site were obtained from near the curb of the asphalt-paved driveway (Fig. 2). The SP9 sampling site was relatively closer to the old incinerator than the SP10 sampling site. Field survey results indicated that the level of surface soil contamination decreased with increasing distance from the incinerator (Tables 11 and 12). The soil core samples had a well developed Ap horizon with dark grayish brown color, fine granular structure, but had small limestone gravels throughout the cores. The top 3-in. segments of the SP9 cores were used for this investigation.

Table 7. Radionuclide concentrations in SP5 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP5-1-A	0-4	1.51E+02	4.94	6.92E+00	5.92	4.59	0.354	3.24E+00	14.27	3.65E-01	12.84
SP5-1-B	4-9	1.32E+02	6.38	6.63E+00	6.90	5.02	0.472	7.88E+00	7.55	1.83E-01	22.87
SP5-2-A	9-12	6.68E+00	12.45	3.22E-01	11.68	4.82	0.822	1.36E+01	1.33	1.13E-02	MDA
SP5-2-B	12-16	6.75E+00	14.93	3.22E-01	16.55	4.77	1.064	1.42E+01	1.61	1.60E-02	MDA

^a²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

^b²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

^c⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

^d¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

Table 8. Radionuclide concentrations in SP6 soil core samples at varying depths

ID	Depth (in.)	$^{238}\text{U}^a$ (pCi/g)	1 sigma % error	$^{235}\text{U}^b$ (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	$^{40}\text{K}^c$ (pCi/g)	1 sigma % error	$^{137}\text{Cs}^d$ (pCi/g)	1 sigma % error
SP6-1-A	0-4	9.30E+01	10.64	3.33E+00	16.78	3.58	0.71	1.23E+01	8.97	3.54E-01	24.90
SP6-1-B	4-8	1.35E+01	32.65	7.01E-01	23.95	5.20	2.11	1.21E+01	5.92	3.36E-02	MDA
SP6-1-C	8-12	8.84E+00	14.59	1.63E-01	27.88	1.84	0.58	1.32E+01	1.79	1.47E-02	MDA
SP6-2-A	12-16	2.86E+00	20.72	1.26E-01	22.81	4.41	1.36	1.36E+01	1.44	1.08E-02	MDA
SP6-2-B	16-20	2.85E+00	26.22	1.83E-01	18.62	6.44	2.07	1.32E+01	1.44	1.08E-02	MDA
SP6-2-C	20-24	2.13E+00	33.12	1.59E-01	19.48	7.48	2.87	1.34E+01	1.43	1.12E-02	MDA

^{238}U activity based on the 100.00-keV (0.92%) gamma energy line for $^{234\text{m}}\text{Pa}$.

^{235}U activity based on the 143.77-keV (10.50%) gamma energy line.

^{40}K activity based on the 1460.73-keV (10.70%) gamma energy line.

^{137}Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ^{235}U .
Normal atmospheric fallout from ^{137}Cs is around 1 pCi/g.

Table 9. Radionuclide concentrations in SP7 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP7-1-A	0-3	3.57E+02	4.19	1.84E+01	4.18	5.15	0.305	9.39E+00	10.53	3.03E-01	20.04
SP7-1-B	3-6	1.51E+02	6.98	7.91E+00	7.22	5.24	0.526	1.37E+01	8.12	7.69E-02	43.91
SP7-1-C	6-9	7.40E+01	8.34	3.10E+00	8.34	4.18	0.494	1.44E+01	5.37	5.66E-02	MDA
SP7-1-D	9-12	5.37E+01	9.84	2.77E+00	10.30	5.17	0.736	1.22E+01	5.68	3.38E-02	MDA
SP7-2-A	0-2	3.75E+02	1.09	1.78E+01	1.24	4.73	0.078	1.01E+01	2.97	3.01E-01	6.29
SP7-2-B	2-5	1.46E+02	1.54	7.39E+00	1.63	5.08	0.114	1.22E+01	1.81	1.51E-01	7.30
SP7-2-C	5-8	7.72E+01	1.96	3.63E+00	2.05	4.70	0.133	1.20E+01	1.65	1.39E-02	MDA
SP7-2-D	8-11	4.61E+01	5.12	2.05E+00	4.28	4.44	0.296	1.31E+01	2.66	1.95E-02	MDA
SP7-3-A	0-3	6.00E+02	3.28	2.91E+01	3.39	4.85	0.229	1.21E+01	12.50	7.36E-01	13.73

²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ratio is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

Table 10. Radionuclide concentrations in SP8 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1-sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP8-1-A	0-2	7.60E+01	3.31	4.51E+00	2.41	5.94	0.243	1.52E+01	2.09	8.47E-01	2.96
SP8-1-B	2-5	3.02E+01	16.94	1.91E+00	13.26	6.34	1.364	1.38E+01	6.17	3.82E-01	12.79
SP8-1-C	5-8	1.41E+01	8.29	7.15E-01	7.34	5.08	0.566	1.49E+01	1.60	1.38E-01	7.89
SP8-1-D	8-11	5.92E+00	20.61	1.70E-01	30.04	2.87	1.045	1.60E+01	1.49	5.66E-02	MDA
SP8-2-A	0-3	6.30E+01	4.48	3.81E+00	3.67	6.05	0.350	1.43E+01	2.72	7.44E-01	3.49
SP8-2-B	3-6	1.38E+01	11.41	7.21E-01	10.68	5.20	0.813	1.40E+01	2.55	1.05E-01	10.63
SP8-2-C	6-9	6.64E+00	28.34	2.95E-01	24.18	4.44	1.656	1.47E+01	2.49	6.15E-02	29.64
SP8-2-D	9-12	7.05E+00	26.29	4.07E-01	24.25	5.78	2.066	1.55E+01	3.21	3.63E-02	MDA
SP8-3-A	0-3	6.85E+01	13.21	4.09E+00	13.39	5.97	1.122	1.42E+01	8.03	9.02E-01	10.08

²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

Table 11. Radionuclide concentrations in SP9 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP9-1-A	0-3	1.68E+03	1.80	8.17E+01	1.85	4.87	0.126	1.12E+01	9.86	6.31E-01	26.45
SP9-1-B	3-6	4.67E+02	3.68	2.53E+01	3.43	5.43	0.273	8.37E+00	11.69	4.61E-01	24.30
SP9-1-C	6-9	1.67E+02	6.13	8.90E+00	6.58	5.33	0.480	1.24E+01	7.15	1.33E-01	42.06
SP9-1-D	9-11	3.95E+01	3.60	2.10E+00	3.76	5.33	0.277	1.14E+01	1.99	4.86E-02	20.21
SP9-3-A	0-3	1.93E+03	1.84	9.42E+01	1.66	4.88	0.121	1.32E+01	10.04	7.70E-01	21.99
SP9-3-B	3-6	3.69E+02	3.62	1.97E+01	3.99	5.33	0.287	1.10E+01	7.11	2.58E-01	29.04
SP9-3-C	6-9	7.35E+01	9.33	3.41E+00	9.44	4.63	0.615	1.17E+01	6.60	7.74E-02	40.91
SP9-3-D	9-12	1.47E+01	22.36	1.56E+00	19.77	10.59	3.161	1.01E+01	7.45	7.80E-02	34.46

^a²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

^b²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

^c⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

^d¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

000033

2321

Table 12. Radionuclide concentrations in SP10 soil core samples at varying depths

ID	Depth (in.)	²³⁸ U ^a (pCi/g)	1 sigma % error	²³⁵ U ^b (pCi/g)	1 sigma % error	%5/8 ActRatio	1 sigma % error	⁴⁰ K ^c (pCi/g)	1 sigma % error	¹³⁷ Cs ^d (pCi/g)	1 sigma % error
SP10-1-A	0-2	3.26E+02	4.51	1.71E+01	4.76	5.24	0.343	1.08E+01	9.47	5.12E-01	17.49
SP10-1-B	2-5	2.58E+02	4.24	1.31E+01	5.13	5.05	0.336	1.07E+01	8.15	3.85E-01	21.53
SP10-1-C	5-8	5.25E+01	10.39	2.52E+00	10.86	4.79	0.721	1.17E+01	6.28	1.38E-01	24.46
SP10-1-D	8-10	3.13E+01	4.65	1.68E+00	4.26	5.36	0.338	1.31E+01	1.77	8.40E-02	10.59
SP10-2-A	0-2	2.20E+02	1.43	1.16E+01	1.21	5.27	0.099	1.18E+01	2.19	4.89E-01	4.23
SP10-2-B	2-4	1.24E+02	2.14	6.14E+00	2.08	4.95	0.148	1.13E+01	2.27	3.48E-01	4.59
SP10-2-C	4-7	4.40E+01	3.40	2.20E+00	3.55	5.00	0.246	1.23E+01	1.71	1.37E-01	8.03
SP10-2-D	7-10	2.00E+01	3.86	9.67E-01	4.18	4.84	0.275	7.89E+00	1.53	5.99E-02	8.87

^a²³⁸U activity based on the 100.00-keV (0.92%) gamma energy line for ^{234m}Pa.

^b²³⁵U activity based on the 143.77-keV (10.50%) gamma energy line.

^c⁴⁰K activity based on the 1460.73-keV (10.70%) gamma energy line.

^d¹³⁷Cs activity based on the 661.65-keV (85.10%) gamma energy line.

Note: The natural 5/8 activity ration is 4.6% for 0.72% ²³⁵U.
Normal atmospheric fallout from ¹³⁷Cs is around 1 pCi/g.

Soil Particle Size Distributions

The results of the particle size fractionation are in Table 13. The >2-mm gravel fractions were probably overestimated because of surface coating of finer materials on coarse fragments. The SP2-2 and SP2-3 samples had a similar particle size distribution pattern. Gravel fractions constituted about 56 to 59% by weight and other fractions were less than 20%. The SP4 sample had 8% gravel, 20% sand, 54% silt, and 18% clay that, if the gravel fraction is discounted, is similar to the surface texture of local loess soils (see reference soils). The SP5-1 sample had 54% gravel, 15% sand, 25% silt, and 16% clay, with the high gravel content reflecting the presence of the coarse slag materials in the soil surface layer. The surface soil of the SP8 sample had 34% sand, 47% silt, and 19% clay. This soil had a similar texture as the reference soils, indicating that the sampling area had been minimally disturbed (Table 13). The surface soil of the SP9 cores had 13% gravel, 21% sand, 53% silt, and 13% clay. The mixing with limestone gravel altered the soil texture somewhat, but the texture of the SP9 soil was similar to other less disturbed soils inside the plant.

The results of the particle size distribution suggest that (1) soils inside the plant boundary were highly disturbed from past construction activities; (2) most of the coarse fragments (>2 mm) were limestone that was used as fill, cover, and road construction materials; (3) the presence of limestone fragment is reflected in the relatively high pH of the surface soils in the sampling areas (Table 14); and (4) the weak alkaline pH and carbonate mineral availability would contribute to a high uranium concentration in perched water zones in the soils. The amounts of uranium, cation, and anions dissolved during size separation were given in Table 15.

Uranium Distribution with Particle Size in Soils

Analytical results of uranium in the soils (Table 13) were expressed in concentration of each fraction ($\mu\text{g/g}$) as well as contribution of each fraction to total soil concentration (%).

Table 13. Soil particle size distribution and uranium distribution with particle size of the soil samples

Soil Location	Size Fraction (mm)	% Particle Size	U Conc. ($\mu\text{g/g}$)	Particle Size Conc. ($\mu\text{g/g}$)	% U Contribution by Size Fraction
SP2-2-ABC	>4	48	134	64.32	8.21
SP2-2-ABC	4 - 2	11	297	32.67	4.17
SP2-2-ABC	2 - 0.053	14	1070	149.8	19.12
SP2-2-ABC	0.053 - 0.002	19	1990	378.1	48.27
SP2-2-ABC	<0.002	8	1980	158.4	20.22
SP2-3-ABC	>4	46	197	90.62	2.52
SP2-3-ABC	4 - 2	10	207	20.7	0.58
SP2-3-ABC	2 - 0.053	12	13900	1668	46.47
SP2-3-ABC	0.053 - 0.002	18	5290	952.2	26.53
SP2-3-ABC	<0.002	14	6130	858.2	23.91
SP4-1A/2A	>2	8	50.1	4.008	0.06
SP4-1A/2A	2 - 0.053	20	15900	3180	47.84
SP4-1A/2A	0.053 - 0.002	54	4560	2462.4	37.04
SP4-1A/2A	<0.002	18	5560	1000.8	15.06
SP5-1-AB	>4	39	159	62.01	13.61
SP5-1-AB	4 - 2	15	924	138.6	30.42
SP5-1-AB	2 - 0.053	25	653	163.25	35.83
SP5-1-AB	0.053 - 0.002	16	386	61.76	13.56
SP5-1-AB	<0.002	5	600	30	6.58
SP8-1A/2A/3A	2 - 0.053	34	283	96.22	46.78
SP8-1A/2A/3A	0.053 - 0.002	47	125	58.75	28.56
SP8-1A/2A/3A	<0.002	19	267	50.73	24.66
SP9-1A/3A	>2	13	791	102.83	2.71
SP9-1A/3A	2 - 0.053	21	8770	1841.7	48.47
SP9-1A/3A	0.053 - 0.002	53	2220	1176.6	30.97
SP9-1A/3A	<0.002	13	5220	678.6	17.86

Table 14. pH measurements of contaminated and reference soils

Sample Name	pH	Sample Name	pH
SP1-1-A	8.2	SP6-1-A	8.2
SP1-1-B	8.3	SP6-1-B	8.6
SP1-1-C	8.1	SP6-1-C	8.3
SP1-3-A	8.2	SP6-2-A	8.4
SP1-3-B	8.1	SP6-2-B	8.2
SP1-3-C	7.8	SP6-2-C	8.4
SP2-1-A	7.8	SP7-1-A	8.2
SP2-2-A	8.1	SP7-1-B	8.2
SP2-3-A	8.0	SP7-1-C	8.2
		SP7-1-D	8.2
SP3-1-A	8.3	SP8-1-A	6.1
SP3-1-B	8.4	SP8-1-B	6.9
SP3-2-A	8.1	SP8-1-C	7.3
SP3-2-B	8.4	SP8-1-D	7.6
SP3-2-C	8.5		
SP3-2-D	8.3	SP9-1-A	7.2
SP3-3-A	8.2	SP9-3-B	7.8
SP3-3-B	8.1	SP9-3-C	8.0
SP3-3-C	8.1	SP9-3-D	8.2
SP3-3-D	8.0		
SP4-1-A	7.9	SP10-1-A	7.3
SP4-2-A	8.3	SP10-1-B	7.6
SP4-2-B	8.4	SP10-1-C	8.0
SP4-2-C	8.5	SP10-1-D	8.3
SP4-2-D	8.2		
SP4-3-A	8.2	Henshaw	6.3
SP4-3-B	8.2	Fincastle	5.4
SP4-3-C	8.3		
SP4-3-D	7.6		
SP5-1-A	8.3		
SP5-2-A	7.9		
SP5-2-B	7.9		

Table 15. Uranium, cations, and anions dissolved during particle size separation

	Uranium ($\mu\text{g/g}$)	Al ($\mu\text{g/g}$)	Ba ($\mu\text{g/g}$)	Ca ($\mu\text{g/g}$)	Fe ($\mu\text{g/g}$)	Mg ($\mu\text{g/g}$)	Mn ($\mu\text{g/g}$)	P ($\mu\text{g/g}$)	Na ($\mu\text{g/g}$)	Sr ($\mu\text{g/g}$)	Alk ($\mu\text{g/g}$)	F ($\mu\text{g/g}$)	SO ₄ ($\mu\text{g/g}$)
SP2-2-ABC	17.2	0	9.3	1404	0	128	0	11.6	26.1	3.7	812	0	0
SP2-3-ABC	214.9	0	12.5	1344	0	161	0	0	45.5	3.4	1330	126	0
SP4-1A/2A	145.3	0	13.2	1402	0	168	0	0	47.5	3.5	1241	113	0
SP5-1-AB	5.83	0	4.5	896	0	144	0	0	21.6	2.8	1080	0	295
SP8-1A/2A/3A*	0.61	43.0	12.9	1063	39.4	222	1.7	0	14.3	2.3	100	0	0
SP9-1A/3A*	1.22	38.6	8.1	934	22.5	174	0	32.2	16.1	1.5	354	0	0
Fincastle Soil*	0.40	0	28.1	756	0	144	8.3	0	0	2.5	0	0	0
Henshaw Soil*	0.01	0	15.0	807	0	147	0	0	110	2.6	0	0	0

*Uranium values for these soils are preliminary.

Note: A "0" value means not detected.

The gravel fractions (limestone fragments) of the SP2-2 and SP2-3 samples had low uranium contamination and their contribution to total uranium was less than 13%. The sand fraction of SP2-2 had a moderate level of contamination and contribution to total uranium concentration, but the sand fraction of SP2-3 contributed about 46% to the total soil uranium concentration, although the soil sample had only 12% sand by weight. On the other hand, the silt fraction of SP2-2 was the largest contributor of uranium to the soil, although the sample had only 19% silt by weight. The clay fractions of the both sample had a moderate contribution (20 to 24%).

The SP4 sample had the highest contamination among the samples (0.665%). The uranium contribution by the gravel fraction was minimal (0.06%). The sand fraction had the highest concentration (15.9 mg/g) as well as the highest contribution (48%) to the total uranium in the soil. The silt and clay fraction had a considerable amount of uranium but their contributions were lower than the sand fraction (Table 13).

The uranium distribution pattern of the SP5 sample was distinctively different from other samples. The SP5 sample had a large amount of gravel fractions (54%) and the gravel fractions were the major uranium contributor (44%). The second largest contributor was the sand fraction (36%). In other samples, gravel fractions were minor uranium contributors regardless of the amount of limestone gravel in the soil.

The uranium concentration of the SP8 sample was the lowest among the samples, but it was still much higher than the background level. The sand and clay fractions had a higher concentration than that of the silt fraction. The sand fraction contributed about 47% of the total uranium in the soil. The uranium distribution in the SP9 size fractions was similar to other samples such as SP4 and SP8. The sand fraction had a higher uranium concentration and was a major contributor although it was a minor size fraction of the soil.

The results of the uranium distribution with size fractions indicate that (1) the majority of uranium in the soils was as individual discrete particles or as smaller particles cemented to silt, sand, and gravel fractions rather than an adsorbed form on clay minerals;

(2) the dominant size fraction for uranium contribution would be sand for the SP2-3, SP4, SP8, and SP9, silt for the SP2-2, and gravel for the SP5; and (3) separation and removal of a particular size range would not help a great deal for the waste volume reduction for most samples except the separation of the gravel fraction from the SP2 soil.

Microscopic Analyses

Polished sections of soil aggregates embedded in epoxy resin were examined by SEM and EDX. Most of the uranium containing clumps in the SP2 samples consisted of aggregates of fine silt or clay particles. The uranium containing aggregates were composed of silicon, aluminum, calcium, phosphorous, and iron (Plate 1 and 2). In the micrographs, individual uranium particles are brighter than silicate minerals. The SP4 specimen contained a wide variety of sizes and shapes of particles containing uranium (Plates 3 and 4). Some particles were composed entirely of uranium and others had iron, calcium, and/or silicon.

The slag material in the SP5 samples was the major source of uranium. Uranium occurred as a coating on the slag surface or as an occluded form in the calcium silicate matrix (Plates 5, 6, and 7). Silicon and calcium were the major elemental components in the slag matrix (Plate 5). The presence of occluded uranium in the slag would cause problems for the development of a decontamination strategy. The uranium-rich particles were much less abundant in the SP8 specimen. Most of the uranium particles were aggregated with silicate minerals. Phosphorous was commonly associated with the uranium particles (Plate 8). Numerous uranium particles having different morphology and composition were observed from the SP9 specimen (Plates 9, 10, and 11). Calcium and phosphorous were detected in some of the particles. Other particles had only uranium (possibly uranium oxides). Some of the particles were mixed with silicate particles as an aggregate form and others were a form of grain separated from a silicate matrix.

Heavy Liquid Separation

The soil samples (<2 mm) were treated with CBD solution to remove amorphous iron coatings. The CBD treatment removed some of the uranium associated with coatings and precipitated/adsorbed on the surface of soil particles (see Chemical Leaching Experiment section). The removal of iron coatings and disaggregation of soil clays would assist in evaluating the effectiveness of heavy liquid separation and mineralogical analysis of the soils. The results of the heavy liquid separation are given in Table 16.

Table 16. Weight and uranium distribution of sand-size fractions after heavy liquid density separation in lithium metatungstate solution with a density 2.8 g/mL.

Sample	Fraction	Weight Distribution (%)	Uranium Distribution (%)
SP2-2-ABC	Floating	83	49
	Heavy	17	51
SP2-3-ABC	Floating	79	64
	Heavy	21	36
SP4-1A/2A	Floating	63	30
	Heavy	37	70
SP9-1A/3A	Floating	82	49
	Heavy	18	51

The data presented in the table were the first separation test results for the sand-sized fraction in the lithium metatungstate solution at density 2.8 g/mL. The separation

procedure such as solid to liquid ratio, tube size, and liquid density will be modified for future experiments. The weight percent of heavy (sink) fractions varied from 17 to 37% and the percent of contribution by the heavy fraction to total concentration varied from 36 to 70%. The results indicated that the heavy liquid density separation is a relatively effective method for the removal of uranium containing particles from the soils. However, the method may not be practical for massive amounts of soils in an engineering scale. Therefore, a new separation technology based on the difference of particle density should be developed for this ID program. Readjustment of the procedure after microscopic and X-ray diffraction examination of the heavy fraction would improve the separation. The heavy liquid separation will be continued for silt and clay fractions and the results will be reported in the final report of this Phase 1 Characterization project.

Mineralogical Analyses

For mineralogical analysis, clay fractions of the soil samples were separated from sand and silt fractions after CBD treatment by the centrifugation method of Jackson (1975). The six contaminated soils had very similar clay mineral compositions. The X-ray diffraction patterns (XRD) of the contaminated soil clays were remarkably similar to those of the Fincastle and Henshaw clay samples. The XRD patterns of clay fractions from SP4-1A/2A and Henshaw soils were shown in Fig. 4 as examples. The XRD after magnesium and ethylene glycerol solvation showed very weak 18-, 14-, 10-, 7-, and 3.34-Å peaks with other second order peaks. After heating K saturated samples to 550°C, the 7-Å peak disappeared. The XRD results indicated that the clay fractions were composed of smectite, vermiculite, mica, kaolinite, and quartz. Swelling clays consisting of smectite and vermiculite were a minor component in the soils. Since clay minerals control many chemical and physical properties of soil, clay mineral composition could influence treatability of soils, dewatering after soil washing, and disposal of secondary waste. Since the clay mineral composition of all core samples was similar, these soil clays should respond about the same to decontamination treatments.

SP4-1A/2A, <2 μm Fraction

HENSHAW, <2 μm Fraction

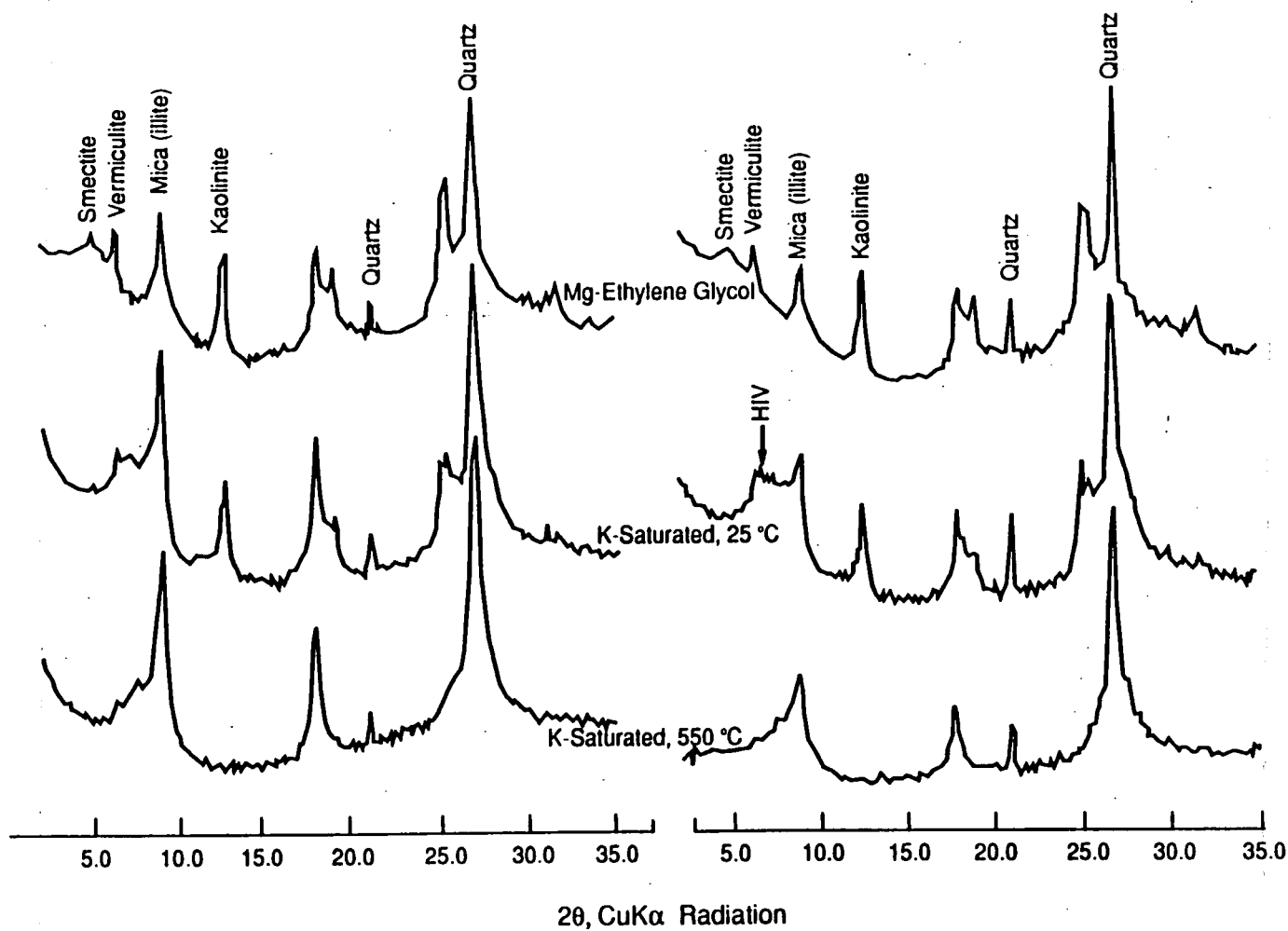


Figure 4. XRD of clay fractions of SP4-1A/2A and Henshaw soils

Chemical Leaching Experiments

The chemical leaching experiments were conducted to provide general information to the Treatability Task Group. The mild acid (0.1 N nitric acid) treatment would remove uranium precipitates on the soil mineral surfaces and dissolve some uranium associated with carbonates. However, the mineral acid treatment was not effective because the contaminated soils contained too many limestone gravels (Table 17). The hydroxylamine hydrochloride treatment would remove uranium associated with manganese coatings in the contaminated soils. The nominal effectiveness of this treatment suggested that extraction of manganese would not have an effect on uranium leaching. Sodium hypochlorite is an effective oxidant for a reduced form of uranium. Oxidation of the reduced uranium would promote leaching by inducing complexation with soil carbonates. The experimental results showed some positive but minimal effects. EDTA is a well known chelating ligand for metals. The treatment was very effective for the SP4 soil sample but was not effective for the other soil samples. CBD treatment is a standard method for removing amorphous sesquioxides (iron and aluminum) in soils. As expected, considerable amounts of uranium (10 to 30%) were removed by this treatment. The bicarbonate and citrate in the solution could be complexed with uranium when the sesquioxides were dissolved by the treatment. The citric acid and ammonium carbonate treatments were the most effective, particularly for the SP4, for removing uranium. Uranium in the soils might be dissolved from solid phase by the citric acid and then complexed as citrate. The excess carbonates in the ammonium carbonate solution would promote complexation of uranium in the contaminated soils.

SUMMARY AND RECOMMENDATIONS

The Phase I soil sampling and characterization task was established by the Characterization Group to obtain basic information related to soil properties and the nature of uranium contamination. The results of this preliminary investigation were intended for (1) the selection of contaminated soils for use in treatability studies, (2) the selection of a field demonstration area or areas within Operable unit 3, and (3) the preliminary screening of integrated decontamination technology.

Table 17. Percent extractable uranium and total uranium concentrations in the soil samples

Sample ID	pH	Extractants							Total U ($\mu\text{g/g}$)
		0.1 N HNO_3 1.7	0.1M $\text{NH}_4\text{OH}\cdot\text{HCl}$ 2.4	2% Citric Acid 2.5	0.1M EDTA 4.6	2% $(\text{NH}_4)_2\text{CO}_3$ 9.5	5% NaOCl 11.7	CBD 8.5	
SP2-2-ABC		2.32	2.09	32.04	1.77	21.66	7.82	28.83	2220
SP2-3-ABC		3.77	2.59	32.05	3.18	20.20	7.11	29.76	2970
SP4-1A/2A		12.43	23.51	74.86	45.41	48.78	25.27	31.28	5920
SP5-1-AB		8.43	6.12	20.45	8.96	9.79	5.58	11.27	575
SP8-1A/2A/3A		14.71	0.11	32.36	9.04	47.14	34.57	27.68	224
SP9-1A/3A		6.31	0.03	43.55	5.44	14.12	6.06	25.10	5290

Ten soil sampling locations were selected from five different areas: Plant 2/3 area, storage Pad Area, Decontamination Pad/Drum Baling Area, Plant 6 Area, and Incinerator Area (Figs. 1 and 2). After examining general soil properties such as pH, texture, morphology, and radionuclide survey by gamma scanning, five sampling locations (SP2, SP4, SP5, SP8, and SP9) representing each area were selected for detailed characterization. Surface soil samples with varying depths were prepared for characterization because of a higher uranium contamination, except the SP2 location. Two subsurface soil samples were selected from SP2 core, one from 10 to 20 in. (SP2-2) and from 20 to 31 in. (SP2-3). Two reference soils representing undisturbed plant area soils were also collected from about 1 to 2 miles west of Fernald Operation Site (Fig. 3).

Selection of Contaminated Soils for Use in Treatability Studies and Demonstration Areas

Three areas, Plant 2/3 represented by SP1 and SP2 samples, Plant 1 Drum Storage Area representing by SP3 and SP4 samples, and Incinerator Area representing SP9 and SP10 samples, have potential to be used for treatability studies. The other two areas, Decontamination Pad/Drum Baling Area and Plant 6 area investigated, are not good candidates because the uranium concentration is too low for technology evaluation and/or the depth of contamination is too shallow for excavation without mixing with uncontaminated soils. In addition, the presence of contaminated slag material in the Decontamination Pad/Drum Baling Area would be a negative element for the effectiveness of decontamination demonstration.

Plant 2/3 Area: The history of contamination, depth of contamination, and nature of contamination suggested that the area was contaminated by aqueous uranium waste. However, the characterization results indicated that silt and sand size fractions were the major uranium contributor in the soil. The microscopic analysis and density separation also indicated that some of the uranium in the soil was in particulate form having a density higher than 2.8 g/mL. Leaching experiment showed relatively low uranium extractability although the uranium particulates appeared as amorphous precipitate forms (Plates 1 and 2). EDX analysis showed that most uranium-containing particles also had calcium and phosphorous as elemental components. Since the contaminated zone had a fairly high

content of slightly contaminated but easily cleaned limestone gravel, a combination of extraction and physical size separation treatment would achieve more than 60% of waste volume reduction.

Plant 1 Drum Storage Area: The area was suspected to be contaminated by uranium product spills. As expected, the nature of the contamination is very complex. More than 80% of uranium was associated with sand and silt fractions comprising about 74% of the soil. However, the citric acid and ammonium carbonate solutions were able to extract about 75 and 49% of uranium from the soil, respectively (Table 17). The uranium particles were associated with iron, calcium, phosphorous, and silicon (Plate 3). Others had only uranium (oxide or metallic ?) (Plate 4). Density-based separation was also successful in isolating uranium-containing heavy particles from the sand fraction (Table 16). This is an excellent candidate area for demonstration of decontamination technologies based on chemical extraction and density separation.

Incinerator Area: This area was suspected to have been contaminated by airborne uranium material. As expected, uranium-containing particles having different composition, shape, and sizes were in the soil samples collected from the area (Plates 9, 10, and 11). Chemical leaching of uranium by citric acid was moderately effective (43%) but was not very effective by ammonium carbonate solution (14%) (Table 17). Heavy liquid density separation was also moderately effective (Table 16). It will be a very difficult engineering challenge to remove uranium or reduce waste volume because the most abundant size fractions (sand and silt) had the most uranium (79% of total uranium) in the soil.

Preliminary Screening of Integrated Technology

Preliminary leaching test results indicated that (1) citric acid dissolution/complexation and carbonate complexation were the most effective methods for removing adsorbed or leachable uranium in the contaminated soils, (2) oxidant and inorganic mineral acid were the least effective leachate solutions, (3) the amount of extracted uranium did not correlate with either total amounts of uranium in soils or in clay fractions. Therefore, chemical extraction alone cannot be expected to accomplish soil remediation. Particle size

distribution and uranium distribution with particle size fractions suggested that simple size separation would not always be effective for waste volume reduction. However, density-based separation showed some degree of success for removing uranium-containing particles. Therefore, the integrated technology to be developed should be based on both chemical extraction and physical separation technologies.

Characterization Group: (1) need more sampling and characterization to verify the preliminary investigation results (e.g., soil samples underneath structures); (2) should investigate on-going geochemical processes to evaluate decontamination impacts; (3) should establish sample preparation and analytical methods as a part of the QA procedure; and (4) should prepare evaluation protocol for the treatment effectiveness and secondary waste disposal technology.

Treatability Group: (1) technologies should be able to remove both particulate form and leachable forms of uranium in these soils, (2) proposed technologies should recycle leaching solution after removal of uranium, (3) should prepare several decontamination schemes reflecting the area specific conditions, and (4) decontamination products to be returned to the site should not include a carbonate source.

Excavation Group: (1) need development of a depth sensing remote control device to remove the contaminated surface soils (<8 in.).

Risk Assessment and Regulation Group: (1) need early establishment of a lower limit of uranium concentration for candidate soils and upper limit for treated soil to be returned to the field.

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APPENDIX

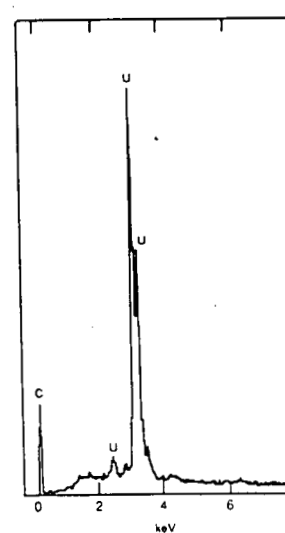
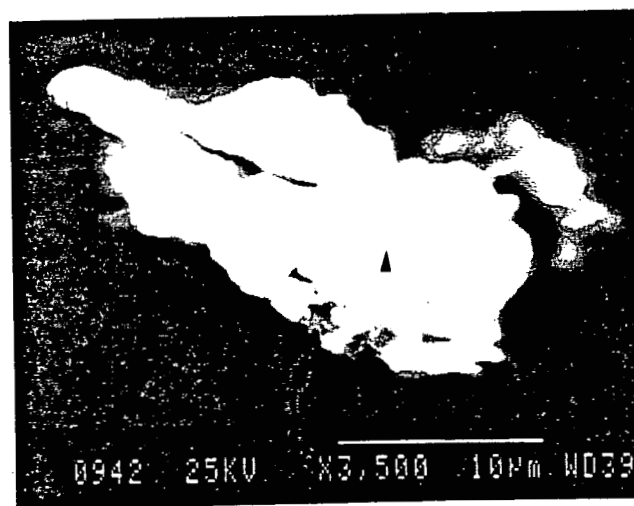
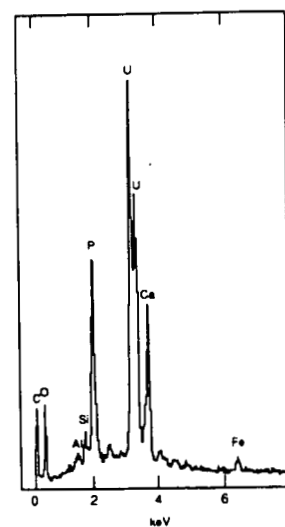
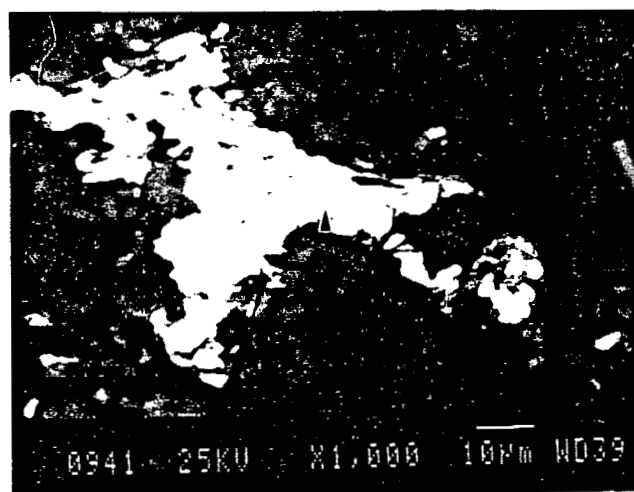
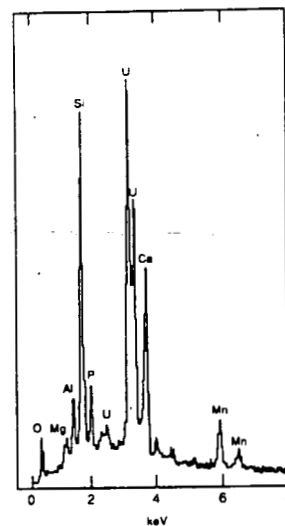
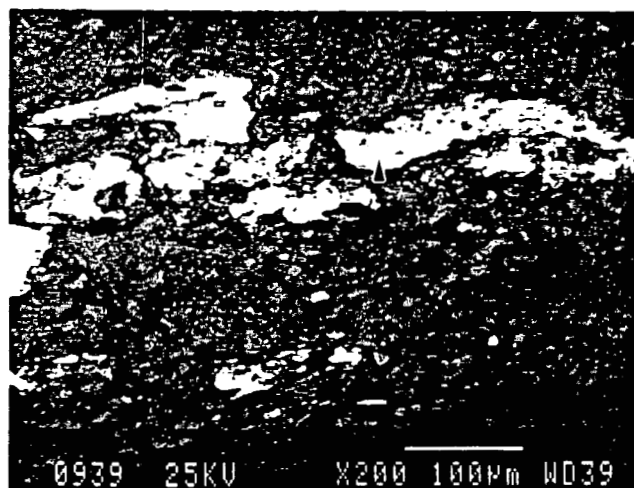


Plate 1. SEM and EDX of uranium-containing particles from SP2 soil sample.

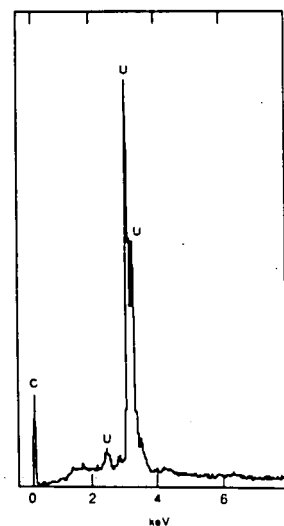
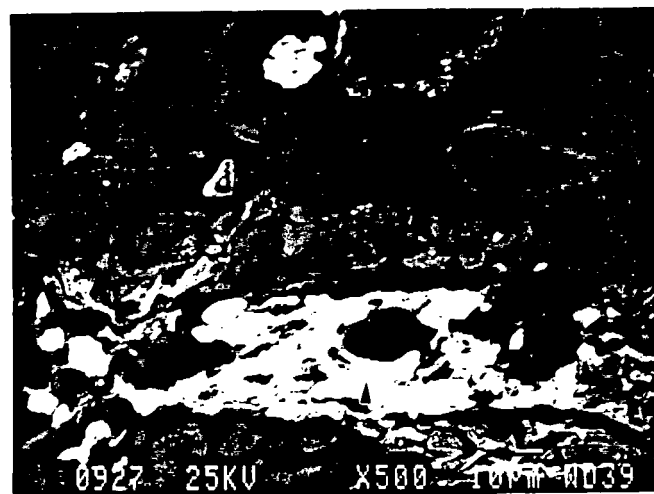
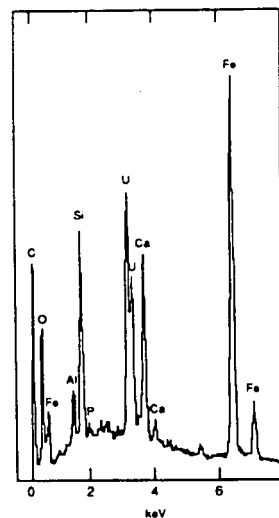
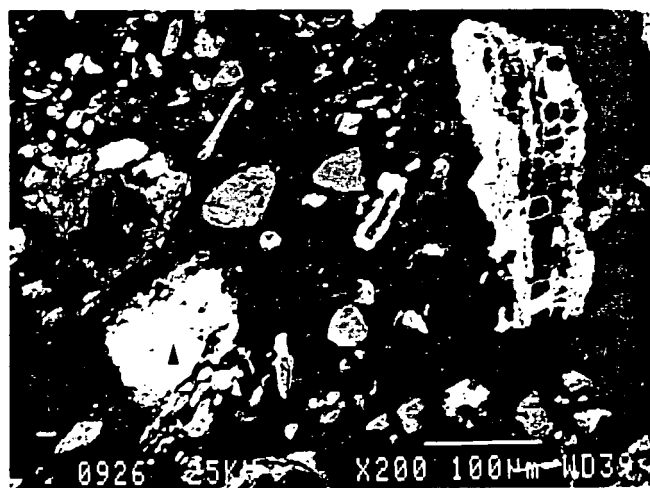
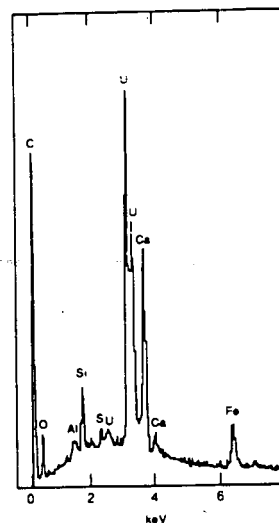
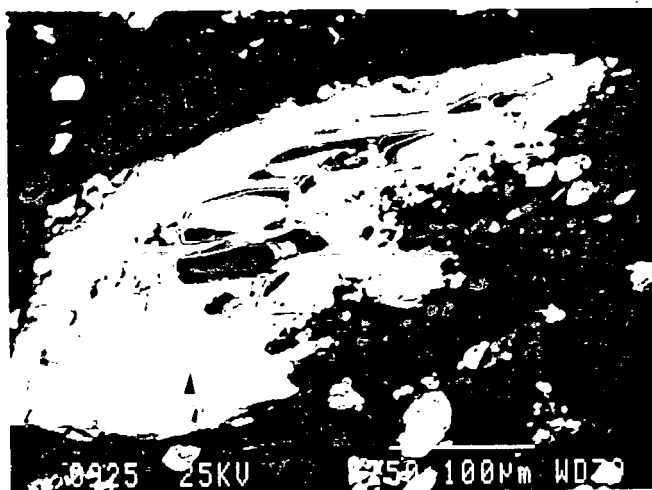


Plate 2. SEM and EDX of uranium-containing particles from SP4 soil sample.

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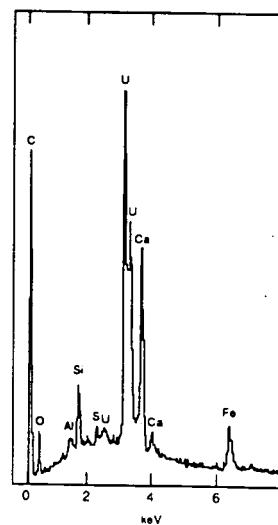
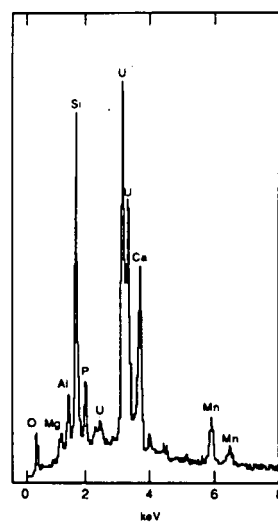
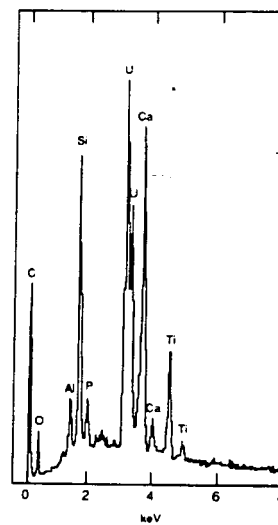
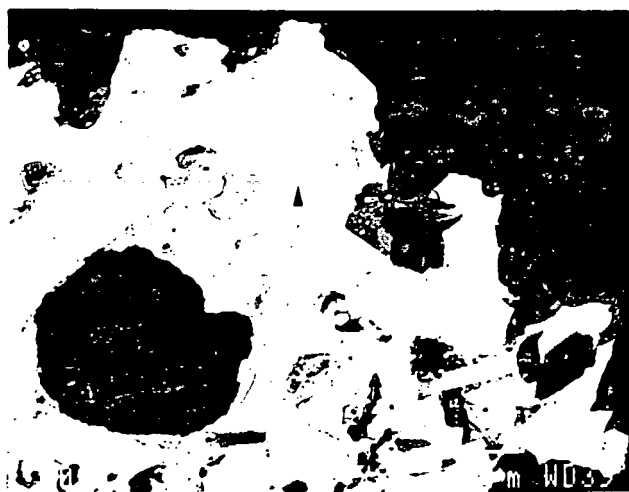


Plate 3. SEM and EDX of uranium-containing particles from SP5 soil sample.

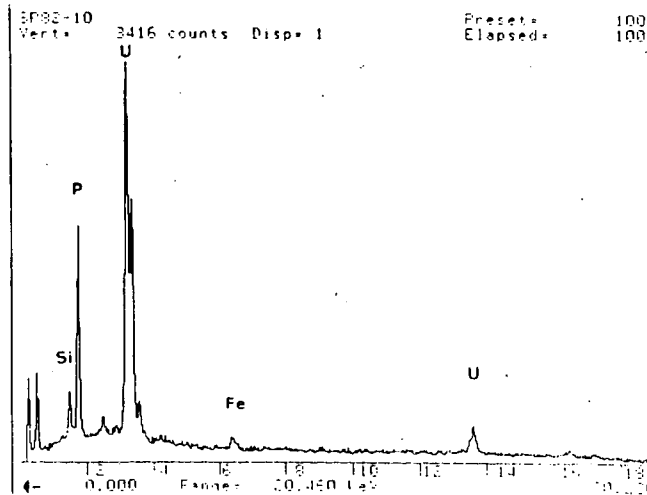
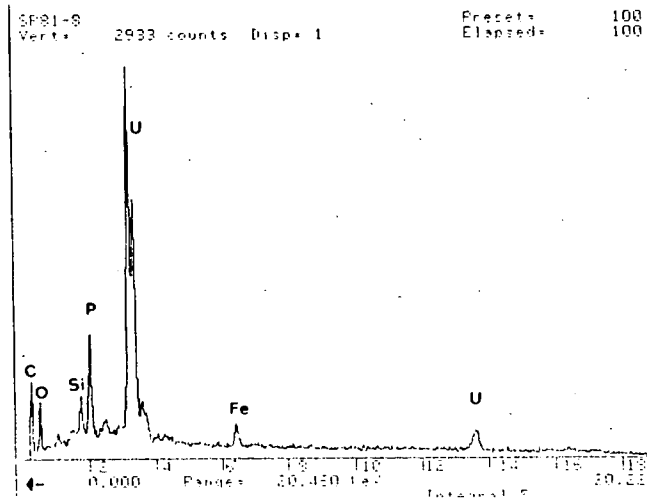


Plate 4. SEM and EDX of uranium-containing particles from SP8 soil sample.

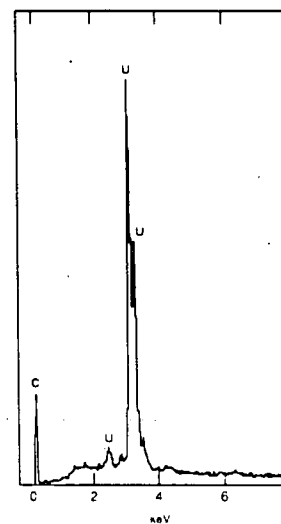
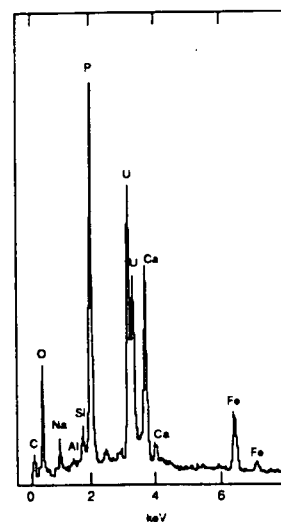
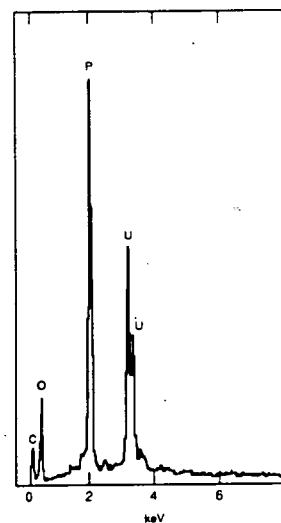
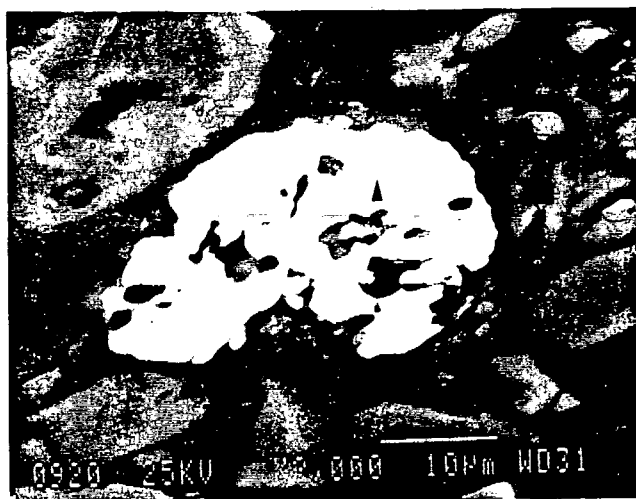


Plate 5. SEM and EDX of uranium-containing particles from SP9 soil sample.

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